

Appl. No. 10/604,664  
Amdt. Dated 12/23/05  
Reply to Office action of 10/11/2005

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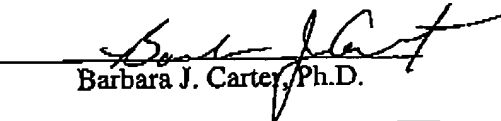
JUL 18 2006

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Ji, J.	Att'y Docket:	3071/103
Serial No:	10/604,664	Confirmation No.:	1663
Date Filed:	August 8, 2003	Art Unit:	1723
Cust. No.:	02101	Examiner:	Fortuna, A.M.
Invention:	DEFECT FREE COMPOSITE MEMBRANES, METHOD FOR PRODUCING SAID MEMBRANES AND USE OF THE SAME		

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DECLARATION OF JIANG JI, PH.D. IN SUPPORT OF  
APPLICANT'S RESPONSE  
[37 C.F.R. § 1.132]

Dear Sir:

In support of the accompanying response to the Office Action mailed October 11, 2005 in the above-reference matter, I hereby declare as follows:

I. My name is Jiang Ji, Ph.D. I am the inventor of the subject matter of the above patent application and am an inventor or co-inventor on a number of other patents and pending patent applications involving membrane technology, including U.S. Patent Nos. 6,596,167 and 6,890,435 and US Application Serial Nos. 10/604,664 and 10/857,531. I have a Ph.D. in Organic/Polymer Chemistry from McMaster University, Hamilton,

Ontario, Canada and I am an internationally known scientist in the field of membrane science and technology and am an author or co-author of a number of publications involving membrane technology, including,

- (i) J. Ji, and M. Mehta, Mathematical Model for the Formation of Thin-Film Composite Hollow Fiber and Tubular Membranes by Interfacial Polymerization, J. Memb. Sci., 192, 41-54, (2001).
- (ii) J. Ji, J. M. Dickson, R. F. Childs, and B. E. McCarry, Mathematical Model for the Formation of Thin-Film Composite Membranes by Interfacial Polymerization: Porous and Dense Films, Macromolecules, 33, 624-633, (2000).
- (iii) J. Ji, B. J. Trunsinski, R. F. Childs, J. M. Dickson, and B. E. McCarry, Fabrication of Thin-Film Composite Membranes with Pendant, Photoreactive Diazoketone Functionality, Journal of Applied Polymer Science, 64, 2381-2398, (1997).
- (iv) J. Ji, R. F. Childs, M. Mehta, Mathematical Model for Encapsulation by Interfacial Polymerization, J. Memb. Sci., 192, 55-70, (2001).
- (v) J. Ji, M. Fei, G. Fan, J. Chen, G. Li, and W. Xiu, The Nucleation of Organic Additives in Membrane Casting Solution (I), The Theory and Direct Evidence, Desalination, 85, 297 -320, (1992).
- (vi) J. Ji, and J. Chen, Study on Interaction between PBIL Membranes and Organic Solutes, Desalination, 78, 389 - 396, (1990).
- (vii) J. Ji, M. Sun, M. Fei, and J. Chen, Study on the Interaction between Membranes and Organic Solutes by the HPLC Method, Desalination, 71, 107 -126, (1989).  
Heteropoly Acid, Chemical Abstract, 108, 111705b (1988), Chemical Journal of Chinese Universities, 6, 165, (1987).

My further credentials are set forth in my Curriculum Vitae, which is attached as Exhibit A hereto.

2. I have read the action of October 1, 2005. This declaration is provided to distinguish the presently claimed membranes and methods of preparing and using such

membranes from the cited art, and explain why the presently claimed invention is novel and non-obvious over the cited art.

3. I have carefully reviewed your comments and the cited patents, and I realize that it is necessary to make some clarifications. I am aware of the prior arts you cited, they may appear to be related to my invention, but they do not provide teaching that will lead to my invention. In contrast, my invention has discovered novel membranes and novel methods for making said novel membranes to overcome the problems that the prior art was unable to solve or did not encounter at the time. My invention has significantly advanced the art of membrane fabrication processes and has produced novel membranes that show much more advanced properties than the prior art.

4. The following is my detailed response to the Examiner's comments in the pending Office Action of October 3, 2005. First, I would like to explain the claimed invention from a chemical reaction perspective.

5. Firstly, the chemical compositions of the membranes of the present invention are different from those of prior art, in particular, patent '039. Under the conditions utilized in the present invention, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate), a key ingredient of the membrane formulation in Examples 1, 2 and 3 of the present invention, reacts with each other catalyzed by aluminum chloride, another key ingredient of the membrane formulation, to form crosslinked three dimensional network, which entangle with poly(vinylidene fluoride) (PVDF) network to strengthen the membrane in the present invention. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form crosslinked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) network having O-Al-O linkages. In this reaction, aluminum chloride plays a role

of both reactant and catalyst. Poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) also reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and O-Al-O bridge to make the membrane permanently bond to polyester braid through covalent bonding. The above reactions of the present invention are totally different from those disclosed in U.S. Patent 5,914,039, where both base and acid are needed in order to get PVDF reacted with polyvinyl alcohol. Thus, the present invention is more advanced than the art disclosed in patent '039.

6. Secondly, the polymeric particles, such as 70 micrometer poly(acrylonitrile-co-methacrylonitrile) particles and crosslinked polyvinylpyrrolidone particles, as key components of the membranes of the present invention render the membranes with much high flux than the calcined  $\alpha$ -alumina particles in patents '039 and '044. The pure water permeability as high as 227 gfd/psi is obtained from the present invention, which is more than 4 times higher than the highest permeability of 50 gfd/psi disclosed in column 12, line 2 of patent '039.

7. Thirdly, the adhesive is covalently bonded to both the braid and the membrane, i.e., the adhesive layer is sandwiched between the membrane and braid by covalent bonding as depicted in Fig. 2 of the present invention to form an adhesive reinforced composite membrane. This is clearly different from Fig. 3 of patent '039, where a single layer membrane made from the same polymer is inaccurately or incorrectly described as four layers, 35, 36, 37 and 38. The further clarification is given in the answers to your questions in claim rejection section.

8. *Claim Rejections-35 USC§ 102(b)* The Examiner asserts that "[c]laims 1-6, 8, 9 are rejected under 35 U.S.C. 102 (b) as being anticipated by Mahendran et al (US

5,914,039) (hereinafter '039)....." (see Office Action, p. 3). In response to the above comment, I have carefully examined patent '039 and found that '039 does not teach the process about how to make defect free composite membrane nor the defect free composite membranes claimed in the present invention.

9. Fig. 3 of patent '039 shows a sketch of membrane cross section, it has multiple regions with different porosity. Although the sketch in Fig. 3 of patent '039 looks somewhat similar to the sketch in Fig. 2 of the present invention, actually it is totally different after careful examination and comparison. In fact, in Fig. 3 of '039, elements 35, 36, 37, 38 are formed from the same polymer by coagulation; they are simply detailed porous structures formed from the same membrane casting solution as shown by a scanning electron photomicrograph in Fig. 4 of '039, the detail description is given in column 8, lines 21-39 of '039. Let me emphasize on this point by quoting column 8 lines 28-33 of '039: "... as evident in great detail in the photomicrograph FIG. 4. The skin is very thin, dense layer of polymer formed as dope contact the coagulant. By reason of the manner in which the skin and each layer is formed from the same polymer, the layers have, in a radially inward direction from under the skin to the braided yarn 39 which defines the bore 32, progressively larger pores." This means that these layers, 35, 36, 37, 38 of '039 are formed from a single membrane casting solution in a single coagulation process at the same time, there is no difference in chemical composition between regions having different pore size and pore size distribution. The sketch shown in Fig. 3 of '039 is an incorrect representation of the true membrane structure shown in Figs. 4 and 5 of '039. Furthermore, '039 does not teach using covalent bonding between braid and coating layer to make a defect free membrane.

10. In contrast to the teaching of '039, the three layers shown in Fig. 2 of the present invention are made by coating two solutions in two sequential steps in a single spinneret shown in Fig. 1 of the present invention. The first coating 22 covers the rough surface of the braid 20 and provides smooth surface for second coating 21. On one hand, the first coating 22 provides covalent bonding with the braid 20 to anchor the coating 22 onto the braid 20 permanently; on the other hand, the first coating 22 also provides covalent bonding with the second coating 21 to form a composite membrane reinforced by covalent bonding within the adhesive layer; within the membrane layer; and between the braid and adhesive layer; as well as between the adhesive and membrane layers. Thus, the composite membranes of the present invention are not only structurally (physically) different, but also chemically different from those disclosed in patent '039.

11. Patent '039 does not reveal membrane defect and adhesion problems between the braid and membrane nor the impact of braid physical structure on membrane performance, but US Pat. No. 6,354,444 (hereinafter patent '444), which is a continuation-in-part of patent '039, issued to the same group of inventors does indeed reveal the problems associated with membrane adhesion, braid physical structure, and pin-hole defects in columns 1 lines 25-67 and column 2 lines 1-36. A method to address these issues is disclosed by emphasis on using different physical structures of braids, Fig. 1 A-C, column 2 lines 49-67, column 3 lines 1-35 and claims 1-11 of '444.

12. In the present invention, these critical problems alluded to in the previous paragraph, in particular, membrane defects and insufficient membrane adhesion to the support, have been solved using chemical reaction approach and sequential multiple-layer coating in a single spinneret. The novel approach of the present invention has not been

taught by the teaching of '039 nor '444, although a complex of PVDF with calcined  $\alpha$ -alumina particles and polyvinyl alcohol are used to make braid support membranes. '039 and '444 do not teach using covalent bonding and multiple coatings to strengthen adhesion and to eliminate membrane defects. Instead, '444 chooses the physical approach using different braid structure to improve membrane adhesion, this physical approach of '444 is far less effective than the chemical approach of the present invention. Thus, the present invention provides more advanced membranes than the prior art in terms of membrane integrity, selectivity and flux. The pure water permeability as high as 227 gfd/psi is obtained from the present invention, which is more than 4 times higher than the highest permeability of 50 gfd/psi disclosed in column 12, line 2 of patent '039.

13. The Examiner asserts that "Adding other polymeric additives is also disclosed, e.g. polymeric derived salts; sulfonated polysulfone (column 7, lines 58-65)." However, all of the polymeric additives described in column 7, lines 58-65 of patent '039 are water soluble, and so they will be eventually washed out of the membrane matrix during separation/filtering processes. Therefore, the impact of these water soluble polymeric additives on membrane performance is temporary; as a result, the membrane performance of '039 becomes worse and worse over time due to the loss of hydrophilic additives.

14. In contrast, the hydrophilic polymeric additives, 70 micrometer poly(acrylonitrile-co-methacrylonitrile) particles and crosslinked polyvinylpyrrolidone particles; and poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate), are not soluble in water, so they are permanently embedded inside the membrane; the crosslinking reaction of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) catalyzed by aluminum chloride make such an embedment even stronger. Thus, the membranes of the present invention have long

lasting hydrophilic components and excellent hydrophilicity, and thus, the membranes of the present invention are more advanced than those disclosed in patent '039.

15. The Examiner alleges that "*Claim 2 is also disclosed (column 8, lines 30-34).*"

But, as discussed above, the chemical composition of the membranes of the present invention is different from those disclosed by patent '039; thus, the membranes of the present invention show much more advanced separation performance than those disclosed by '039 in terms of flux, selectivity and integrity.

16. The second half of the claim 2 of the present invention, i.e. "The membrane of claim 1, wherein said middle layer and said outside barrier layer are formed from.....different coating solutions." is clearly different from that described in column 8, line 23-34 of '039, where so called "layers" are actually different porous regions of the same single layer membrane formed from the same polymer (column 8, lines 31-32). In other words, the '039 authors incorrectly or inaccurately described the different porous regions of a materially same single layer membrane, which real structure is shown in Figs. 4 and 5 of '039, as a multiple-layer membrane in Fig. 3 of patent '039.

17. In contrast to '039, the membrane claimed in the second half of claim 2 of the present invention is a true materially different multiple-layer membrane, each layer has distinct chemical composition; the said middle layer is an adhesive, the said outside barrier layer is made of dope I consisting of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate), aluminum chloride, 70 micrometer particles of poly(acrylonitrile-co-methacrylonitrile), polyvinylpyrrolidone, and poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP). In addition, aluminum chloride also reacts with



poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form crosslinked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) network having O-Al-O linkages.

18. The membrane of the present invention is schematically illustrated in Fig. 2, which is a true multiple-layer membrane. The polyester braid provides mechanical support; PVDF-HFP entangled with three dimensional network of crosslinked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) forms a barrier layer to provide selective separation; and middle adhesive layer formed from specially formulated adhesive, chemically bonds the outer barrier layer and inner braid support together to form a reinforced composite membrane. The middle adhesive layer of the present invention is different from the conventional adhesive in that the middle adhesive layer of the present invention is permeable to fluid, while the conventional adhesive is not permeable to fluid, it is normally used to seal membrane when making a membrane cartridge.

19. The first half of the claim 2 of the present invention, i.e. "The membrane of claim 1, wherein said middle layer and said outside barrier layer are formed from the same coating solution" is also different from that described in column 8, line 3-34 of patent '039 in that the chemical composition of the present invention is different from that of '039. In the present invention, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) is used as a key ingredient of the membrane, which reacts with each other catalyzed by aluminum chloride to form a crosslinked three dimensional network through covalent bonding of acetyl bond linkage. The crosslinked three dimensional network of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) entangles with PVDF-HFP network to strengthen the membrane in the present invention.

20. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form O-Al-O bridge to crosslink multiple poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) macromolecules together. In this reaction, aluminum chloride plays a role of both reactant and catalyst. More importantly, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and oxygen-aluminum-oxygen bridge to make the membrane permanently attach to polyester braid. In this reaction, aluminum chloride again plays a role of both reactant and catalyst.

21. Therefore, the defect free composite membranes claimed in claims 1 and 2 are totally different from those disclosed in patent '039 in terms of both chemical composition and physical structure. Thus, the membranes of the present invention have much more advanced features than those disclosed by the prior art of '039.

22. With respect to the Examiner's assertion "RE: claim 3, the middle layer is formed from organic polymer, e.g. hydrophilic PVDF (column 7, second paragraph)", there are at least two major differences between the claim 3 of the present invention and what is described in column 7, second paragraph of patent '039.

23. (1) *The claim 3 of the present invention emphasizes on the middle layer, "which has excellent compatibility between the support and the barrier layers to bond them together."* The prior art described in column 7, second paragraph does not mention to bond the membrane with the braid support at all. In other words, patent '039 does not teach to bond the membrane with the support. As pointed out above, patent '039 does not reveal membrane defect and adhesion problems (delamination problem) between the braid and membrane nor the impact of braid physical structure on membrane

performance, but US Pat. No. 6,354,444 (hereinafter patent '444) issued to the same group of inventors does indeed reveal the problems associated with poor membrane adhesion, i.e. membrane delamination, braid physical structure, and pin-hole defects in columns 1 lines 25-67 and column 2 lines 1-36.

24. A method to address these issues is disclosed by emphasis on using different physical structures of braids, Fig. 1 A-C, column 2 lines 49-67, column 3 lines 1- 35 and claims 1-11 of '444. In contrast to the teaching of '039, claim 3 of the present invention describe the bonding membrane with the support by adhesives and other organic and inorganic materials which have bonding capacity similar to expoxy, polyurethane, silicone and other adhesives.

25. *(2) Claim 3 of the present invention describes the middle layer of the membrane is selected from the group of consisting of expoxy, polyurethane, silicone and other adhesives, including other organic and inorganic materials which have bonding capacity similar to adhesives. The prior art described in column 7, second paragraph does not teach using adhesive at all, instead describes PVDF/ $\alpha$ -Aluminum complex, which does not provide bonding between the membrane and support as revealed by the same group of inventor in a subsequent US Patent No. 6,354,444.*

26. Therefore, the prior art described in column 7, second paragraph of patent '039 does not teach claim 3 of the present invention. In contrast, claim 3 of the present invention claims much more effective composition of matters than the prior art of '039 to bond chemically and physically membrane and support together.

27. With respect to the Examiner's assertion that the "Limitation of claims 4-6 are further disclosed in patent '039 (see column 7, lines 1-36)", membrane performance is

determined by a membrane's chemical composition and physical structure. In column 7, lines 1-36, of patent '039, only PVDF is used as a major component, which forms complex with  $\alpha$ -Al, (PVDF/ $\alpha$ -Al complex). In contrast, the major component of the membrane of the present invention is selected from a group that is far broader than PVDF, as described in claim 4, although PVDF is included. Such a group of polymers offers unique and dramatically different performance from those disclosed in '039, e.g. poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) is stable under pH=14 condition, while PVDF is only stable under pH=12 or less.

28. Thus, the membrane made of poly(vinylidene-co-hexafluoropropylene) of the present invention is more stable than those PVDF membranes disclosed in patent '039. On the other hand, the membranes made of poly(acrylonitrile-co-butadiene-co-styrene), poly(vinylidene-co-acrylonitrile), poly(vinylidene chloride-co-acrylonitrile-co-methacrylate) of the present invention are more resistant to oil and oily waste water than those PVDF membranes of patent '039.

29. The minor component of the membrane of the present invention as claimed in claim 5 is also significantly different from those used in '039. For example, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) can be easily crosslined to form 3-dimensional networks under the conditions used in the present invention to make membrane more stable than those of '039. Even poly(vinyl alcohol), sulfonated polysulfone and cellulose acetate used in the present invention react with aluminum chloride or undergo the different crosslinking reactions catalyzed by aluminum chloride. That is totally different from the membranes of '039.

30. The polymeric particles of claim 6 are totally different from the calcined  $\alpha$ -alumina of '039. Calcined  $\alpha$ -alumina are very porous materials and are normally used as absorbents. As a result, the membranes of patent '039 having  $\alpha$ -alumina as additives are easy to foul by absorbing the suspended particles and other fouling materials from solution. In contrast, the water insoluble but hydrophilic polymeric particles as membrane additives of the present invention do not absorb those fouling materials easily; thus, the membranes of the present invention experience much less fouling than the membranes of '039.

31. Furthermore, calcined  $\alpha$ -alumina particles precipitate out of the membrane solution in one or two days time period. That means that the concentration of the calcined  $\alpha$ -alumina in a membrane casting solution changes with time in patent '039, which results in a larger variation in membrane performance. In contrast, the polymeric particles used in the present invention do not precipitate and so the membrane casting solution is very stable over time. Such a stability is very important to a large scale of membrane production.

32. Regarding the Examiner's comment "Re: claims 7-9, the membrane in tubular or hollow fiber shape is disclosed (column 1, lines 11-26, column 11, lines 39-54, claim 1)", the tubular and hollow fiber membranes are not new, they existed even before patent '039. What is new in the present invention is that, as discussed above, the chemical compositions of the membranes of the current invention are significantly different from those disclosed by patent '039. Thus, the membranes of the present invention show much more advanced performance than those disclosed by '039, in terms of flux, selectivity and integrity. The proper combination of multiple components and processing conditions,

under which the membrane is formed, is very important to determine membrane characteristics.

33. Regarding the Examiner's comment "Re: claim 12, the claimed membrane properties are disclosed in column 10, lines 45-56).", in column 10, lines 45-56 of patent '039, the membranes described have 500 ft<sup>2</sup> area, the highest specific flux is 36.6 GFD/psi, and no burst pressure is described. In contrast, claim 12 of the present invention claims a membrane, which has a burst pressure of 10-500 psi regardless the membrane area; a pure water flux up to about 500 gfd/psi, which is much higher than 36.6 GFD/psi as described by '039; and, retention of poly(ethylene oxide) molecular weight marker having an average molecular weight of 200,000 daltons. The retention is not mentioned at all in column 10, lines 45-56 of patent '039. The novelty and inventiveness of the present invention to eliminate membrane defect and to overcome membrane delamination problem by double layers coated one on top of another in sequence in a single spinneret; and by covalent bonding between the support and coating layers is acknowledged by the Written Opinion of the International Searching Authority for the corresponding PCT application (PCT/US04/25374), a copy of which is attached herewith as Exhibit B.

34. *Claim Rejections-35 USC § 103(a)* Regarding the Examiner's comment: "4. Claim 10-11 is rejected ...", coating a sphere is not new. What is new in the present invention is coating a sphere with multiple reactive coating layers to give a unique spherical *defect free* membrane, which has a chemical composition and separation properties similar to its hollow fiber and flat sheet counterparts.

35. In patent '039, the calcined alumina particle size (<5 micrometer) is much smaller than the thickness of the film. The particles are used as film fillers, and are suspended in the casting solution. This is different from coating a sphere. When coating a sphere, the thickness of the coating layer is normally much smaller than the diameter of the sphere. The volume ratio of coating layer to coated particle is extremely small in the present invention, thus the properties associated with this ratio is quite different from that of patent '039, where the volume ratio of film layer to film particles is huge, and the thickness of the film is about 20 times larger than the size of calcined alumina particle. US Pat. No. 5,766,473 discloses the coating of thermally formed polyethylene and polypropylene microfiltration membranes with tactic hydrophilic poly(vinyl alcohol). On a micro scale, polyethylene and polypropylene membrane have a microporous structure, the fibrous polyethylene and polypropylene are connected by randomly shaped knots to form a three dimensional network. Tactic poly(vinyl alcohol) covers these fibrous knots to form a shell on the outer surface. Enzymes as oxygen scavenger are loaded onto the poly(vinyl alcohol) shell. On a macro scale, the membranes of patent '473 have flat sheet geometry.

36. The major differences between the present invention and patents '473 and '039 are as follows: In the present invention, there are three different scenarios:

37. (1) for a spherical support, which has reactive groups, such as esters, the support is first coated by a reactive casting solution, which reacts with the support to form covalent bonding to permanently bond the coating layer to the support; such covalently bonded first coating layer is coated with a second reactive layer that reacts with the first coating layer to form a defect free composite membrane with covalent bonding and

crosslinking to strengthen the defect free composite membrane. As described above, in the present invention, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) is used as a key ingredient of the membrane, which reacts with each other catalyzed by aluminum chloride to form a crosslinked three dimensional network through covalent bonding of acetyl bond linkage and O-Al-O linkages. The crosslinked three dimensional network of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) entangles with PVDF network to strengthen the membrane in the present invention.

38. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form O-Al-O bridge to crosslink multiple poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) macromolecules together. In this reaction, aluminum chloride plays a role of both reactant and catalyst. More importantly, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and oxygen-aluminum-oxygen bridge to make the membrane permanently attach to polyester braid. In this reaction, aluminum chloride again plays a role of both reactant and catalyst.

39. (2) For a spherical support, which do not have obvious reactive groups, the support is first coated by a reactive adhesive layer which is not cured or partially cured during coating, the adhesive penetrates into the support. This reactive first coating layer is coated again with a second reactive layer, which reacts with the first coating layer to form a defect free composite membrane strengthened by covalent bonding and crosslinking within and between the layers.

40. (3) For a support, which has reactive groups, such as esters, the support is first coated by a reactive adhesive layer which is not cured or partially cured during coating,



the reactive adhesive reacts with ester group to form covalent, the adhesive also penetrates into the support and reacts with each other to form crosslinked first coating layer which covalently anchored onto the support, which is coated again with a second reactive layer, which reacts with the first coating layer to form a defect free composite membrane strengthened by covalent bonding and crosslinking.

41. Thus, the spherical membranes of the present invention are structurally (physically) and chemically different from those of prior art. The present invention emphasizes using reactive multiple coating layers, one layer on top of another, and covalent bonding and cross linking to make defect free composite membranes which have much higher flux, integrity and selectivity than those membranes disclosed in patents '039 and '473. The membranes disclosed in patent '473 are for oxygen removal, thus do not have liquid separation properties. This is totally different from the present invention. Thus, patent '473 does not provide teaching for the present invention.

42. Regarding the Examiner's comment that "Claims 2 and 3 are rejected under 35 U.S.C. 103 (a)...", careful examination of patent '039 indicates that patent '039 actually discloses a coating solution of the same polymers. In example 1, column 11, line 1 to 36 of patent '039, the detailed formulation of the coating solution of the same polymers and a dope preparation procedure are described. In example 2, column 11 line 42 "The dope formed in Example 1 is fed to a nozzle .....". US Patent No. 6,024,872, which is a division of patent '039, also discloses the detail formulation of coating solution of different polymers and dope preparation procedure.

43. Regarding the Examiner's comment that "Patent '193 teaches a braided support impregnated and coated with a first polymer, coating the support to form a layers on the

braided polymeric material, and further using the coated support to cast a film on a additional polymeric membrane, to increase the thickness of the membrane is disclosed (see entire disclosure, in particular claims 1, 8-11 and 21)", after having carefully examined patent '193 and the entire disclosure, in particular claims 1, 8-11 and 21, I found that it is necessary to clarify the difference between the present invention and patent '193.

44. The major differences between the present invention and patent '193 are as follows: In the present invention, there are three different scenarios:

45. (1) *For a support, which has reactive groups, such as esters, the support is first coated by a reactive casting solution, which reacts with the support to form covalent bonding to permanently bond the coating layer to the support; such covalently bonded first coating layer is coated with a second reactive layer that reacts with the first coating layer to form a defect free composite membrane with covalent bonding and crosslinking to strengthen the defect free composite membrane.*

46. (2) *For a support, which do not have obvious reactive groups, the support is first coated by a reactive adhesive layer which is not cured or partially cured during coating, the adhesive penetrates into the support. The first reactive coating layer is coated again with a second reactive layer, which reacts with the first coating layer to form a defect free composite membrane strengthened by covalent bonding, crosslinking and covalently anchoring onto the support.*

47. (3) *For a support, which has reactive groups, such as esters, the support is first coated by a reactive adhesive layer. Before the first reactive coating layer is cured, it is coated again with a second reactive layer, which reacts with the first coating layer to form*

a defect free composite membrane firmly anchored onto the support by covalent bonding and crosslinking.

48. The membranes of the present invention are also physically and chemically different from those of disclosed in the patents cited by the examiner. In the present invention, the adhesives including epoxy, polyurethane, silicones react with both membrane and support to covalently bond the membrane and support together to form a support reinforced membranes, which are much stronger, much more permeable, and much more selective than those disclosed in prior art. More importantly, the prior art does not provide teaching that will lead to the present invention.

49. The coating processes of the present invention for making defect free composite membranes are much more advanced than those disclosed in prior art, in particular, patents '039 and 193. The coating processes of patent '193 are manual processes. The tubular membranes claimed in patent '193 are prepared by a non-continuous batch mode at very slow coating speed, the longest tube disclosed in '193 is 44 ft long.

50. The coating process of patent '039 are described in column 3 lines 51-64, and in column 11 lines 40-60. A tubular braid is coated with a layer of dope containing calcined  $\alpha$ -alumina particles, polyvinyl alcohol and PVDF at a speed of 40 ft/min, coagulated in water to form a braid support membrane. No ultrasonic wave stirring is used to facilitate phase inversion.

51. In contrast, the membrane of the present invention is prepared by coating a braid with two reactive layers simultaneously in a single spinneret, one layer on top of the other, at a coating speed as high as 100 ft/min or higher. The ultrasonic wave generators are used to provide ultrasonic agitation in coagulation bath to facilitate mass transfer

between cast membrane and coagulation liquid to speed up phase inversion from liquid to solid of the cast membrane. Thus, the membranes are coated at much higher speed in the present invention than prior art. The membranes in the present invention are produced in a continuous mode. The membranes are collected by two interchangeable membrane take-up wheels, when one is full, switching to another, the process can be operated around the clock, 24 hours a day, 7 days a week. The coating processes of the present invention are much more efficient and much more productive those disclosed in the prior art.

52. Thus, the present invention emphasizes on using reactive multiple coating layers, one layer on top of another, covalent bonding and cross linking to make defect free composite membranes which have much higher flux, integrity and selectivity than those membranes disclosed in patent '193.

53. The above discussion clearly shows that the novel membranes and novel processes for making said novel membranes of the present invention are much more advanced than those disclosed in prior art. The prior art does not provide teaching that will lead to the present invention. Thus, even the people who are skilled at art cannot develop of the same membranes and processes of the present invention by following the teaching of the prior art.

54. In conclusion, the membranes of the present invention are different in chemical compositions and physical structures from the membranes disclosed in the prior art. And more importantly, the membranes of the present invention show much higher water flux and much higher burst pressure than the membranes disclosed in the prior art. Some of

the prior arts may appear to be relevant, actually these prior arts do not provide any teaching that will lead to the present invention.

55. I hereby declare that all statements made herein are of my own knowledge and that all statements made on information and belief are true; and further that these statements are being made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully yours,



Jiang Ji  
Tel: (603) 560-2147  
Fax: (603) 458-1483  
Email: [jjjiang@comcast.net](mailto:jjjiang@comcast.net)  
4 Hawk Drive  
Salem, NH 03079

Dated: December 23, 2005

03071/00103 455309.1

**Exhibit A****Jiang Ji, Ph.D.**

Member of American Chemical Society  
Member of North America Membrane Society

**EXPERIENCE****June 2004 – Present**

Staff Scientist  
Amersham Biosciences, GE Healthcare  
General Electric Company  
Westborough, MA 01581

Research and development of cutting edge technology  
and products for purification of cervical cancer vaccine

**Sept. 1998 – May, 2004**

Senior Polymer Scientist  
Koch Membrane Systems Inc., Koch Industries, Inc.  
Wilmington, MA, USA

Research and development of reverse osmosis (RO),  
nanofiltration (NF), ultrafiltration (UF), and  
microfiltration (MF) membranes having different  
geometries, ranging from hollow fiber, tubular to flat  
sheet membranes for water purification, wine filtration,  
juice filtration, and other applications in biotech,  
pharmaceutical, food and chemical industries

**Sept. 1995 - Sept. 1998**

Senior Research Chemist  
Zenon Environmental Inc., ON, CANADA

Research and development of braid reinforced hollow  
fiber membranes and tubular membranes for drinking  
water purification, wastewater treatment and other  
applications in auto, food, biotech, pharmaceutical and  
chemical industries.

**March - Aug. 1989**

Visiting Research Scientist  
University of Oxford, UK

Study of natural phospholipid vesicles/transition metal  
catalysts

Sept. 1988 - Feb. 1989	<b>Visiting Research Scientist</b> University of Ottawa, CANADA  Study on reverse osmosis membranes and nanofiltration membranes
June 1986 - Aug. 1988	<b>Research Scientist</b> Dalian Institute of Chemical Physics Chinese Academy of Sciences  Fundamental research on membrane formation, membrane transport mechanism and interactions between membrane and separated solutes
July 1985 - May 1986	<b>Lecturer</b> Dalian University of Technology Teaching physical chemistry and chemical engineering
<b>EDUCATION</b>	
Sept. 1989 - Aug. 1995	<b>Ph.D. Organic/Polymer Chemistry</b> Department of Chemistry McMaster University, Hamilton, ON, CANADA  Ph.D. Dissertation: Fabrication and Photochemical Surface Modification of Photoreactive Thin-Film Composite Membranes and Model Development for Thin Film Formation by Interfacial Polymerization
Sept. 1982 - July 1985	<b>M.Eng.</b> Department of Chemical Engineering Dalian University of Technology M.Eng. Thesis: Catalytic Characteristics of the Keggin Type of Heteropolyacids
March 1978 - Jan. 1982	<b>B.Eng.</b> Department of Chemical Engineering Dalian University of Technology B.Eng. Thesis: Synthesis of poly(ethylene-co-butadiene) using Ziegler- Natta catalyst and rare earth metal catalysts

Exhibit?

## PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

To: JIANG JI 4 HAWK DRIVE SALEM, NH 03079
--

PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL SEARCH REPORT AND  
THE WRITTEN OPINION OF THE INTERNATIONAL  
SEARCHING AUTHORITY, OR THE DECLARATION

(PCT Rule 44.1)

WORKING COPY

Applicant's or agent's file reference 000037663	Date of mailing (day/month/year)
International application No. PCT/US04/22502	FOR FURTHER ACTION See paragraphs 1 and 4 below
Applicant JI, JIANG	International filing date (day/month/year) 14 July 2004 (14.07.2004)

1. ☒ The applicant is hereby notified that the international search have been established and are transmitted to the applicant and the written opinion of the International Searching Authority.
- Filing of amendments and statements in response to the international search report (see Rule 46):
- When? The time limit for filing the international search report.
- Where? Directly to the International Searching Authority, 1211 Geneva 26.
- For more detailed instructions, see the International Searching Authority's website.
2. ☐ The applicant is hereby notified that the declaration under Article 17(2)(a) to that effect and the written opinion of the International Searching Authority are transmitted to the applicant.
3. ☐ With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:
- ☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.
- ☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.
4. Reminders
- Shortly after the expiration of 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.
- The applicant may submit comments on an informal basis on the written opinion of the International Searching Authority to the International Bureau. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established. These comments would also be made available to the public but not before the expiration of 30 months from the priority date.
- Within 19 months from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later); otherwise, the applicant must, within 20 months from the priority date, perform the prescribed acts for entry into the national phase before those designated Offices.
- In respect of other designated Offices, the time limit of 30 months (or later) will apply even if no demand is filed within 19 months.
- See the Annex to Form PCT/IB/301 and, for details about the applicable time limits, Office by Office, see the PCT Applicant's Guide, Volume II, National Chapters and the WIPO Internet site.

Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (703) 305-3230	Authorized officer Ana M. Fortuna Telephone No. (571) 272-1700
---	--

Form PCT/ISA/220 (January 2004)

(See notes on accompanying sheet)



## PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

To:  
JIANG JI  
4 HAWK DRIVE  
SALEM, NH 03079

## PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL SEARCH REPORT AND  
THE WRITTEN OPINION OF THE INTERNATIONAL  
SEARCHING AUTHORITY, OR THE DECLARATION

(PCT Rule 44.1)

Date of mailing  
(day/month/year)

24 OCT 2004

Applicant's or agent's file reference  
000037663

FOR FURTHER ACTION See paragraphs 1 and 4 below

International application No.  
PCT/US04/22502

International filing date  
(day/month/year) 14 July 2004 (14.07.2004)

Applicant  
JI, JIANG

1. ☒ The applicant is hereby notified that the international search report and the written opinion of the International Searching Authority have been established and are transmitted herewith.

Filing of amendments and statement under Article 19:

The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46):

When? The time limit for filing such amendments is normally two months from the date of transmittal of the international search report.

Where? Directly to the International Bureau of WIPO, 34 chemin des Colombettes  
1211 Geneva 20, Switzerland, Facsimile No.: (41-22) 338.82.70.

For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect and the written opinion of the International Searching Authority are transmitted herewith.

3. ☐ With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.

☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

## 4. Reminders

Shortly after the expiration of 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.

The applicant may submit comments on an informal basis on the written opinion of the International Searching Authority to the International Bureau. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established. These comments would also be made available to the public but not before the expiration of 30 months from the priority date.

Within 19 months from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later); otherwise, the applicant must, within 18 months from the priority date, perform the prescribed acts for entry into the national phase before those designated Offices.

In respect of other designated Offices, the time limit of 30 months (or later) will apply even if no demand is filed within 19 months.

See the Annex to Form PCT/IB/301 and, for details about the applicable time limits, Office by Office, see the PCT Applicant's Guide, Volume II, National Chapters and the WIPO Internet site.

Name and mailing address of the ISA/US

Mail Stop PCT, Attn: ISA/US  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, Virginia 22313-1450  
Facsimile No. (703) 305-3230

Authorized officer

Ana M. Fortuna

Telephone No. (371) 272-1700

Form PCT/ISA/220 (January 2004)

(See notes on accompanying sheet)

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 000037663	<b>FOR FURTHER ACTION</b> <small>see Form PCT/ISA/220 as well as, where applicable, item 5 below.</small>	
International application No. PCT/US04/22502	International filing date (day/month/year) 14 July 2004 (14.07.2004)	(Earliest) Priority Date (day/month/year) 16 July 2003 (16.07.2003)
Applicant JI, JIANG		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 1 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

## 1. Basis of the Report

a. With regard to the language, the international search was carried out on the basis of:



the international application in the language in which it was filed.



a translation of the international application into \_\_\_\_\_, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

b. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the international application, see Box No. I.

2. ☐ Certain claims were found unsearchable (See Box No. II)

3. ☒ Unity of invention is lacking (See Box No. III)

4. With regard to the title,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the abstract,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the drawings,

a. the figure of the drawings to be published with the abstract is Figure No. 2



as suggested by the applicant.



as selected by this Authority, because the applicant failed to suggest a figure.



as selected by this Authority, because this figure better characterizes the invention.

b. ☐ none of the figures is to be published with the abstract.

Form PCT/ISA/210 (first sheet) (April 2005)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

## Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

See USPTO299 (ATTACHED).

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of any additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.: 1-9

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet(2)) (April 2005)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

## Box IV TEXT OF THE ABSTRACT (Continuation of Item 5 of the first sheet)

The technical features mentioned in the abstract do not include a reference sign between parentheses (PCT Rule 8.1(d)).

## NEW ABSTRACT

A defect free semipermeable membrane ((Fig. 2) ) having excellent integrity and high water permeability is provided. Said composite membrane comprises an inside support layer (20) to provide sufficient mechanical strength, and outside barrier layer (21) to provide selective separation and a middle layer (22) to provide both chemical and physical binding between the support and the barrier layers. Three different methods for making said defect free are disclosed. These methods have been successfully utilized to produce high quality coatings and defect free composite membranes, which are independent of chemical composition and physical structure of said support. In the present invention, the ultrasonic sonification is utilized to enhance mass transfer and to speed up the phase inversion process of a membrane casting solution, and to produce a composite membrane at a speed higher than those disclosed in the prior art. Said defect free composite membrane have broad applications, ranging from filtration of fruit juice, wine, and milk to purification of drinking water, and municipal and industrial water.

Form PCT/ISA/210 (continuation of first sheet(3)) (April 2005)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : B01D 61/00, 63/00

US CL : 210/490, 500.27, 500.35, 500.41, 500.36, 500.42; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/45

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 210/490, 500.27, 500.35, 500.41, 500.36, 500.42; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/45

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5,914,039 A (MAHENDRAN et al) 22 June 1999, column 3, lines 52-63, column 7, lines 18-33, column 8, lines 21-39, column 8, lines 30-34, column 7, second paragraph, column 11, lines 39-54, claim 1, and column 10, lines 45-56)	1-6, 8, 9
—		7
Y		
Y	US 5,766,473 A (STROBEL et al) 06 June 1998, column 5, lines 5-61, abstract, column 4, lines 5-20 and last paragraph.	7
Y	US 3,676,193 A (COOPER et al) 11 July 1972, claims 1, 8, -11, and 21, entire disclosure.	2, and 4

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "B" earlier application or patent published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- "Z" document member of the same patent family

Date of the actual completion of the international search

28 September 2005 (28.09.2005)

Date of mailing of the international search report

24 OCT 2005

Name and mailing address of the ISA/US

Mail Stop PCT, Attn: ISA/US  
 Commissioner for Patents  
 P.O. Box 1450  
 Alexandria, Virginia 22313-1450

Facsimile No. (703) 305-3230

Authorized officer

Ana M. Fortuna

Telephone No. (571) 272-1900

Form PCT/ISA/210 (second sheet) (April 2005)

## PATENT COOPERATION TREATY

From the  
INTERNATIONAL SEARCHING AUTHORITYTo:  
JIANG JI  
4 HAWK DRIVE  
SALEM, NH 03079

PCT

WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43bis.1)

Applicant's or agent's file reference		Date of mailing (day/month/year)	
000037663		24 OCT 2005	
FOR FURTHER ACTION		See paragraph 2 below	
International application No.	International filing date (day/month/year)	Priority date (day/month/year)	
PCT/US04/22502	14 July 2004 (14.07.2004)	16 July 2003 (16.07.2003)	
International Patent Classification (IPC) or both national classification and IPC			
IPC(7): B01D 61/00, 63/00 and US Cl.: 210/490, 500.27, 500.35, 500.41, 500.36, 500.42; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/45			
Applicant			
JI, JIANG			

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☒ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

## 2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (703) 305-3230	Date of completion of this opinion 28 September 2005 (28.09.2005)	Authorized officer Ana M. Fortuna Telephone No. (571) 272-1700
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Form PCT/ISA/237 (cover sheet) (April 2005)

WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY

International application No.

PCT/US04/22502

## Box No. I Basis of this opinion

1. With regard to the language, this opinion has been established on the basis of:

- ☒ the international application in the language in which it was filed  
☐ a translation of the international application into \_\_\_\_\_, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

2. With regard to any nucleotide and/or amino acid sequence disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:

a. type of material

- ☐ a sequence listing  
☐ table(s) related to the sequence listing

b. format of material

- ☐ on paper  
☐ in electronic form

c. time of filing/furnishing

- ☐ contained in the international application as filed.  
☐ filed together with the international application in electronic form.  
☐ furnished subsequently to this Authority for the purposes of search.

3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table(s) relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

4. Additional comments:

Form PCT/ISA/237(Box No. I) (April 2005)

WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY

International application No.

PCT/US04/22502

## Box No. IV Lack of unity of invention

1. ☒ In response to the invitation (Form PCT/ISA/206) to pay additional fees the applicant has, within the applicable time limit:
- ☐ paid additional fees
  - ☐ paid additional fees under protest and, where applicable, the protest fee
  - ☐ paid additional fees under protest but the applicable protest fee was not paid
  - ☒ not paid additional fees
2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose not to invite the applicant to pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rule 13.1, 13.2 and 13.3 is
- ☐ complied with
  - ☒ not complied with for the following reasons:  
See the lack of unity section of the International Search Report (Form PCT/ISA/210)

4. Consequently, this opinion has been established in respect of the following parts of the international application:

- ☐ all parts
- ☒ the parts relating to claims Nos. 1-2

Form PCT/ISA/237 (Box No. IV) (April 2005)



**WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY**

International application No.  
PCT/US04/22502

**Box No. V Reasoned statement under Rule 43 bis.1(a)(i) with regard to novelty, inventive step or industrial applicability, citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Claims 7	YES
	Claims 1-6, 8-9	NO
Inventive step (IS)	Claims NONE	YES
	Claims 1-9	NO
Industrial applicability (IA)	Claims NONE	YES
	Claims NONE	NO

**2. Citations and explanations:**

Claims 1-6, 8-9 novelty under PCT Article 33(2) as being anticipated by Mahendran et al (US 5,914,039)(hereinafter '039).

Patent '039 discloses a membrane including the elements (i)-(iii) as claimed in claim 1 (Fig. 1, elements 39, 39', 37, and 38, column 3, lines 52-63, column 7, lines 18-33, column 8, lines 21-39).

Claim 2 is also disclosed (column 8, lines 30-34).

Re: claim 3, the middle layer is formed from organic polymer, e.g. hydrophilic PVDF (column 7, second paragraph).

Limitations of claims 4-6 are further disclosed in patent '039 (see column 7, lines 1-36).

Re: claims 4-6 and 8, the membrane in tubular or hollow fiber shape is disclosed (column 1, lines 11-26, column 11, lines 39-54, claim 1).

Re: claim 9, the claimed membrane properties are disclosed in column 10, lines 45-56).

Claim 7 lacks an inventive step under PCT Article 33(3) as being obvious over Mahendran et al (5,914,039)(hereinafter 'patent '039) as applied to claim 1 above, and further in view of Strobel et al (5,766,473)(hereinafter patent '473).

Patent '039 discloses film formation of the claimed membrane composition, coating spheres or particles with the particular support is not disclosed. It would have been obvious to one skilled in the art at the time the invention was made to use any support, e.g. flat, tubular, or particles for the membrane depending on the intended use, spherical membrane or particles will be suitable for example infiltration in packed columns or in chromatographic separations. Patent '473 teaches hydrophilic porous particles or spheres including a substrate and a coating of hydrophilic membrane (abstract, column 4, lines 5-20 and last paragraph); the substrate materials are also disclosed (column 5, lines 5-61). It would have been obvious to one skilled in the art at the time the invention was made to use a polymeric substrate with spherical shape, as disclosed in '473, to support the membrane of '039, depending on the desired configuration or intended use. As to claim 11, other configurations, such as tubules (e.g. flat or oval shape tube, it would have been obvious to one skilled in the art at the time the invention was made, and can be achieved by shaping or molding the polymeric support before coating, since the support is flexible, modifications of the membrane diameter shape can be within the knowledge of the skilled artisan.

Claims 2 and 4 lack an inventive step under PCT Article 33(3) as being obvious over Mahendran et al (5,914,039)(hereinafter 'patent '039) as applied to claim 1 above, and further in view of Cooper et al (3,676,193)(hereinafter patent '193).

Patent '039 fails to disclose the coating solutions of different polymers, as claimed in a second embodiment of claim 2. Patent '193 teaches a braided support impregnated and coated with a first polymer, coating the support to form a layer on the braided polymeric material, and further using the coated support to cast a film of an additional polymeric membrane, to increase the thickness of the membrane is disclosed (see entire disclosure, in particular claims 1, 8-11, and 21). Based on the discussed teachings, it would have been obvious to one skilled in the art wishing to have a composite membrane on a braided support with distinct layers of polymer, to use the treated support of '193, which contains a layer of polymer, e.g. polyepoxy, polyurethane, etc (column 8, second paragraph), as support, and further cast the membrane with the hydrophilic membrane of '039, e.g. to provide strength to the membrane, and inherent (as produced by epoxy polymer, as claimed in claim 6) easier binding between the hydrophilic.

Form PCT/ISA/237 (Box No. V) (April 2005)

**CHAPTER I**  
**PCT TELEPHONE MEMORANDUM**  
**FOR**  
**LACK OF UNITY OF INVENTION**

---



PCT No.: PCT/US04/22502

Examiner: Ana M. Fortuna

Attorney spoken to: JI JIANG

Date of call: 28 September 2005

- ☐ Amount of payment approved:
- ☐ Deposit account number to be charged:
- ☐ Attorney elected to pay for ALL additional Inventions
- ☐ Attorney elected to pay only for the additional inventions covered by
- ☐ Group(s):
- encompassing --
- ☐ Claim(s):
- ☒ Attorney elected NOT to pay for any additional Inventions, therefore, only the first claimed invention (Group I) covered by Claim(s) 1-9 has been searched.
- ☒ Attorney was orally advised that there is no right to protest for any group not paid for.
- ☒ Attorney was orally advised that any protest must be filed no later than 1 month from the mailing of the Search Report (PCT/ISA/210).

**Time Limit For Filing A Protest**

Applicant is hereby given 1 month from the mailing date of this Search Report in which to file a protest of the holding of lack of unity of invention. In accordance with PCT Rule 40.2, applicant may protest the holding of lack of unity only with respect to the group(s) paid for.

**Detailed Reasons For Holding Lack of Unity of Invention:**

Please See Continuation Sheet

*Note: A copy of this form must be attached to the Search Report.*

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USPTO/299 (August 1997) B

International application No: PCT/US04/22502

**ATTACHMENT TO CHAPTER I PCT TELEPHONE MEMORANDUM  
FOR  
LACK OF UNITY OF INVENTION**

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**Continuation of Detailed Reasons For Holding Lack of Unity of Invention:**

This application contains the following inventions or groups of inventions, which are not so linked as to form a single general inventive concept under PCT Rule 13.1. In order for all inventions to be examined, the appropriate additional examination fees must be paid.

Group I claim(s) 1-9, drawn to a composite membrane.

Group II, claim(s) 10-14, drawn to a method of making a membrane.

Group II, claim(s) 15, drawn to an apparatus.

Group IV, claim(s) 18, drawn to a process of using the membrane.

The inventions listed as Groups I, II, III, and IV do not relate to a single general inventive concept under PCT Rule 13.1 because, under PCT Rule 13.2, they lack the same or corresponding special technical features for the following reasons: the membrane of group I lack the composition, the control of the coating thickness, the speed of collection and the removal of additive recited in claims of group II. Group II includes the feature "spinneret" and "multiple inlets, which features are not recited or required in the claims of groups I, II, or IV. Group IV does not include any common feature with membrane of group I, the process of making the membrane, or the apparatus of group II, and is just directed to using a membrane for multiple fluid separation alternatively.

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*Note: A copy of this form must be attached to the Search Report.*

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## NOTES TO FORM PCT/ISA/220 (continued)

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:  
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
2. [Where originally there were 15 claims and after amendment of all claims there are 11]:  
"Claims 1 to 15 replaced by amended claims 1 to 11."
3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:  
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or  
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
4. [Where various kinds of amendments are made]:  
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

-Statement under Article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

#### Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments and any accompanying statement, under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the time of filing the amendments (and any statement) with the International Bureau, also file with the International Preliminary Examining Authority a copy of such amendments (and of any statement) and, where required, a translation of such amendments for the procedure before that Authority (see Rules 55.3(a) and 62.2, first sentence). For further information, see the Notes to the demand form (PCT/IPEA/401).

#### Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see the *PCT Applicant's Guide*, Volume II.

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